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Interactive comment on "Effects of the physical state of tropospheric ammonium-sulfate-nitrate particles on global aerosol direct radiative forcing" *by* S. T. Martin et al.

M. Jacobson

jacobson@stanford.edu

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The paper by Martin et al. contains many interesting and potentially useful calculations. However, it overlooks relevant information from a previous paper, Jacobson (2001a) (hereafter J2001), cited in the manuscript but hardly discussed. J2001 examined global solid formation and direct forcing among a broader system than sulfate-nitrate-ammonium. Results from the simulation with a broader system differ from those of Martin et al, yet there is no mention of the relevant results from J2001 nor a discussion of the differences. The purpose of this letter is to bring this information to the attention of the authors so that they may consider it in a revision.

J2001 considered direct forcing among 47 aerosol constituents, including 24 electrolyte solids formed from the ions H+, Na+, K+, Mg2+, Ca2+, NH4+, NO3-, HSO4-, SO42-, Cl-, CO32-, and HCO3-. Equilibrium was calculated in and direct forcing was calculated among 17 size bins for $>$400 solar and thermal-infrared wave intervals. For optical calculations, particles were assumed to contain (if present) a black-carbon core surrounded by a shell, where shell refractive indices were found from partial molar refraction theory. The relative humidities used for the calculations were prognostic instantaneous relative humidities (obtained from global simulations). The relevant conclusion comparable with the present study (as stated in the abstract) was ŞSolid formation in aerosols was found to increase total-aerosol direct forcing by +0.03 to +0.05 W/m2.Ť This change in forcing was $<$ 5% of total (anthropogenic plus natural) aerosol forcing calculated therein. The reasons for this relatively small effect are discussed on P. 1553, paragraph 4 of the paper.

The Martin et al. paper appears to have considered solids forming among a smaller system, H+, NH4+, NO3-, HSO4-, and SO42-. Equilibrium calculations appear to have been solved in one size bin and the results distributed lognormally to 11 bins for optical calculations. The use of a single lognormal mode does not account for the multimodal nature of ambient aerosols. The technique also differs from that in J2001 where equilibrium was solved in each of 17 bins independently (instead of one bin) and optical calculations were then applied to each bin. Solid formation will differ when one bin is solved and the results distributed versus when multiple bins are solved independently. Other differences are that Martin et al. appears not to have solved for the thermal-IR nor included core-shell optical treatment nor treated solution refractive indices.

Based on the above, it appears that the following statements in Martin et al. may be mistaken or incomplete.

(1) Section 1.2. ŞPrevious global modeling treatments of aerosol phase include:..(b) calculation of the effects of US and LS of the hysteresis loop on aerosol direct radiative forcing assuming fixed aerosol chemical composition (usually ammonium sulfate) (ĚJacobson, 2001a).Ť

This statement is incomplete since J2001Šs calculation did not assume fixed aerosol composition (concentration and composition of each constituent varied as a function of season, location, relative humidity, temperature, and size) and composition was not limited to ammonium sulfate (it was calculated among 47 constituents).

(2) Section 1.2. ŞPrevious studies of category b (fixed chemical composition) find DFU,L=15 to 20%.find delta forcing of about 15-20%ĚMetzger et al. (2002) report DFU,=5%. Unfortunately, their results seem anomalous.Ť

This statement does not discuss the DFU,L<5% found in J2001 nor possible reasons for the differences. Such a discussion might include the fact that a larger system was solved in J2001 and a different methodology used (see above) in each case. The use of monthly-averaged (by Martin et al.) instead of prognostic instantaneous relative humidities may also play a role (please also see p. 1553 of J2001) and may support the comment of J. Heintzenberg that Şa prognostic treatment of RH is likely to eliminate most of the reported LS/US contrast in aerosol effects.Ť The response to J. Heintzenberg by Martin et al. that Şsuch work at a level equivalent to our treatment of phase transitions has never been completedŤ cannot be correct because the system treated by Martin et al. does not include most of the species (e.g., K, Ca, Mg, Na, Cl, BC, OM, and soil components) and several of the physical processes (described above) treated in J2001.

(3) Section 6.2. ŞWith the exception of Adams et al. (2001) and Metzer et al. (2002), previous studies exclude nitrate.Ť

J2001 included nitrate and many other species.

(4) Abstract. ŞOn the LS, the sulfate mass budget is 40% solid ammonium sulfate, 12% letovicite, ... The LS nitrate mass budget is 26% solid ammonium nitrate,ĚThe LS ammonium budget is 45% solid ammonium sulfateŤ

This conclusion is based on simulations that ignored solids containing Na+, K+, Mg2+,

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Ca2+, Cl-, CO32-, and HCO3-. J2001, who accounted for such solids, found different results, namely ŞGypsum (CaSO4-2H2O), sal ammoniac (NH4Cl),Ě, and nitrum (KNO3) to be the most common sulfate-, ammonium,Ě, and nitrate-containing solid-phase aerosol constituents, respectively, in the global atmosphere.Ť (abstract). Figure 2 of J2001 shows the globally-averaged vertical distribution of modeled solids. These previous conclusions are not discussed in Martin et al. Even though the conclusions have uncertainty, it is certain that sulfate-, nitrate-, and ammonium-solids other than those simulated by Martin et al form. Gypsum may be the most abundant sulfate-containing solid simply because it can form at an RH of 98% and plenty of calcium exists in soil and sea spray globally, as discussed in J2001. The specification of specific fractions of solids in the abstract of Martin et al. is difficult to support given the certain existence of other solids not simulated.

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